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Non-Coalescence Effects in Microgravity

(NAG 3-1894)

Performance Report for the period

17 June 1996 - 16 June 1997

Submitted to:

Dr. Ray Skarda

Microgravity Fluids Branch, Mail Stop 500/102
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by

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May, 1997

I. Summary

Non-coalescence of two bodies of the same liquid and the suppression of contact between liquid drops and solid surfaces is being studied through a pair of parallel investigations being conducted at the Georgia Institute of Technology and the Microgravity Research and Support (MARS) Center in Naples, Italy. Both non-coalescence and contact suppression are achieved by exploiting the mechanism of thermocapillary convection to drive a lubricating film of surrounding gas (air) into the space between the two liquid free surfaces (non-coalescence) or between the drop free surface and the solid (contact suppression). Experiments performed to date include flow visualization experiments in both axisymmetric and (nearly) two-dimensional geometries and quantitative measurements of film thickness in the contact-suppression case in both geometries.

II. Discussion of Research

The apparatus of the axisymmetric experiments reported by Dell'Aversana, Banavar & Koplik (1996) on the non-coalescence of two liquid drops has been modified to obtain additional data. Specifically, an interferometry system has been devised to interrogate the air film between droplets and the lower droplet is capable of being replaced by a solid surface such as a glass plate to investigate the contact-suppression phenomenon. Interferometry measurements in the latter case are capable of yielding an absolute value of the film thickness. A paper recently submitted to *Physics of Fluids* by the MARS group provides detailed examples of this. A preprint of this paper is included as Exhibit B of the subgrantee's performance report, which appears as the Appendix to this report.

An additional modification to the MARS apparatus is presently being made to allow assessment of the influence of surrounding gas pressure on non-coalescence. Obviously, as the absolute gas pressure tends toward zero, the lubricating gas film will reach a point where it is no longer able to sustain the load associated with droplet deformation. Parametric studies will determine threshold values for different states of surrounding pressure and droplet deformation.

At Georgia Tech, an apparatus has been constructed to examine quasi-two-dimensional non-coalescence and contact suppression. Both effects can be achieved in the two-dimensional case, but the drop sizes are quite small for the silicone oils being employed in these experiments. With the assistance of Pasquale Dell'Aversana of the MARS Center, who visited Georgia Tech in March, an interferometry setup has been constructed for this apparatus as well, permitting film thickness determination as described above for the axisymmetric case. Some preliminary parametric studies have been performed with this apparatus, including an examination of behavior as the load between the two drops is increased. In some cases, the drops are observed to coalesce into a two-dimensional liquid bridge, while in others, the pinned contact line holding the drop to the copper heater gives way, allowing liquid to spill over the side of the heater.

Visualization of the flow field within the two-dimensional drop is difficult, due to the highly curved free surface at the ends of the drops. However, we have been able to employ smoke visualization to investigate the flow in the air immediately adjacent to the drops. The motion consists of a vigorous inflow along the surface of the upper, hot drop toward the gap between the droplets accompanied by an outflow along the surface of the lower, cold drop. In the region just outside the convergence of the two drops, a standing vortex is formed. Flow in the gap between the two drops cannot be observed at the present time.

Finally, in both the two-dimensional and axisymmetric cases, an interesting transition to time-dependent flow is observed under certain conditions which are yet to be quantified. This time-dependence is periodic in nature, reminiscent of the instability of thermocapillary convection in liquid bridges. A study of the stability properties of these states which was originally proposed therefore appears to be in order and will be conducted in a subsequent year of this grant.

III. Reference

Dell'Aversana, P., Banavar, J. R. & Koplik, J. 1996 Suppression of coalescence by shear and temperature gradients. *Physics of Fluids* **8**, 15.

IV. Project Personnel

at Georgia Tech:

G. Paul Neitzel, Professor, Principal Investigator

John C. Nalevanko, Graduate Research Assistant

at the MARS Center:

Luigi Carotenuto, Researcher

Dario Castagnolo, Researcher

Pasquale Dell'Aversana, Researcher

V. Publications and Presentations

Dell'Aversana, P., Tontodonato, V. & Carotenuto, L. 1997 Suppression of coalescence and wetting: the shape of the interstitial film. Submitted to *Physics of Fluids*.

Nalevanko, J. C. 1997 Design of an apparatus for investigation of 2-D liquid drop non-coalescence, M.S. thesis, Georgia Institute of Technology.

Neitzel, G. P. & Dell'Aversana, P. 1997 When liquids stay dry. Invited paper in preparation for publication in *Physics Today*.

Neitzel, G. P. 1997 Non-Coalescence of Liquid Drops and Suppression of Surface Wetting. Invited talk presented at the March Meeting of the American Physical Society, Kansas City, MO, March 17-20.

Castagnolo, D., Dell'Aversana, P., Tontodonato, V. & Neitzel, P. 1997 Features of non-wetting and non-coalescing drops. To be presented at the Joint Xth European and Vith Russian Symposium on Physical Sciences in Microgravity, St. Petersburg, Russia, June 15-21.

Appendix

MARS Center Performance Report

Microgravity Advanced Research and Support Center

Non-Coalescence Effects in Microgravity

Performance Report

for the period

June 17, 1996 through April 16, 1997

Subgrantee's part

Subgrant number:	**E-25-L43-G1**
Subgrantee:	**Microgravity Advanced Research and Support Center (MARS)**
Subgrantee address:	**Via Comunale Tavernola,** **80144, Napoli, Italy** **Phone: +39-81-234-4580** **Fax: +39-81-234-7100**
Principal Investigator:	**Dr. Luigi Carotenuto**
Prime grant number:	**NAG3-1894**
Grantee:	**Georgia Institute of Technology**
Principal Investigator:	**Prof. G. P. Neitzel**
Date:	**16 April 1997**

Luigi Carotenuto

Francesco Laccetti
(General Manager)

Non-Coalescence Effects in Microgravity
Performance Report

The present document reports on the Subgrantee's activities being performed for the first year of a study concerning the phenomenon of stable non-coalescence between liquids[1]. The work is being executed jointly with the Georgia Institute of Technology and will be carried out over a four year period. The activities of the first year are expected to be completed by June 16, 1997. They are now being performed according to what announced in the statement of work [Exhibit A] and completed to about 85%.

Stable non-coalescence is the ability of some liquids to be brought in close proximity, and even squeezed against one another, without merge, as long as certain well defined dynamical conditions of their surfaces are mantained. The reason of such surprising behavior had been tentatively ascribed to an action of the liquid surfaces that, when moving, may be able to drag a film of the surrounding medium between the involved liquids. Such film would exert a lubricating force preventing an actual contact of the liquids and thus their coalescence. The aim of this four year project is to achieve a full understanding of the phenomenon, to give a quantitative description of it, to investigate the phyisical limits of non-coalescing systems, and, possibly, to find some application, terrestrial or exploiting microgravity, where the forces concerned assume more relevance than on ground.

In the course of the period reported, the interpretation of non-coalescence in terms of lubrication theory, which was previously based on preliminary experimental results, has been confirmed by interferometric measurements. A theoretical explanation has been outlined, and numerical simulations are presently being worked out to consider the evolution of the most relevant fluid-dynamic parameters of the problem when the systems are stressed. The study has been extended also to the investigation of non-wetting of solid surfaces. In facts, liquid/solid systems, from one hand, may undergo non-wetting owing to the same lubrication mechanisms producing non-coalescence - so representing a different aspect of the same problem - and, on the other hand, liquid/solid systems have resulted to be more suitable for the study of the lubricating film shapes. It has also been evidenced that non-coalescing systems, even being stable, may undergo unsteadiness. The nature of such unsteadiness is being investigated and some hypotheses have been formulated.

Different interferometric techniques have been set out and adapted to the cases of interest in order to reveal the geometrical features of the thin air film separating non-coalescing liquids or a non-wetting liquid and a solid. The experimental apparatus has been configured to yield also non-wetting by means of thermocapillary convection. The liquid/solid configuration is more suitable to yield information on the film shape by means of interferometry because the presence of a flat interface greatly simplifies the interpretation of the interference patterns. In particular, near field interferometry has been used with such configurations and, shifting the angle of incidence of light, from the consequent variation of the interference order, it has been possible to measure the film absolute thickness, besides of its shape. It has been found that far field fringes are, instead, more appropriate to monitor the geometrical features of highly deformed interfaces; for this reason they have been used to reveal the dynamical characteristics of the lubrication film between a drop and a bath of the same liquid when the system becomes unsteady. For the drop/bath system it has been observed that the onset of unsteady flows occurs when a certain threshold in the relevant thermo-fluid-dynamical parameters is overcome. A beam deflection technique has been used to monitor the oscillation modes in unsteady situations.

Dedicated softwares have been developed both to simulate interference fringe patterns for given 3-D film profiles and to analyze the images containing the information about the oscillations of the drop/bath system.

The results have been discussed in terms of lubrication theory and some counterintuitive behaviors, such as the thickening of the lubricating film as the system is loaded, have been evidenced and explained.

An exhaustive discussion of the new findings achieved in the course of the period concerned, is found in the manuscript entitled "Suppression of coalescence and of wetting: the shape of the interstitial film" by P. Dell'Aversana, V. Tontodonato, and L. Carotenuto [Exhibit B]. This manuscript has been submitted for publication in "Physics of Fluids", acknowledging the NASA support which allowed the execution of the work.

The experimental part to be still performed concerns the measurement of critical temperature conditions for coalescence as a function of pressure in the outer environment. Some of the pressure sensors, needed to perform such measurement, have already been received on loan from Georgia Tech, successfully tested, and integrated in the experimental cell. The remaining parts of the cell are completed, including the liquid injection/suction system, the drop displacement mechanism, and the pressurization device, which have been integrated in a pre-existing structure. The signals from the pressure sensors and from the thermocouples, are read by means of a data acquisition board, after signal conditioning, in an integrated National Instruments SCXI system, which is also able to perform data analysis in real time using a Labview software. A software man-machine interface has been realised in order to monitor temperatures and pressures inside the experimental cell simultaneously.

Some numerical simulations have been carried out. A numerical technique based on the control volume method has been utilized to simulate the thermocapillary motion that develops inside a liquid drop suspended at a solid heating disk. The calculated surface velocity profile has been used as a boundary condition for the simulation of the air film in a deformed channel. An orthogonal coordinate transformation based on the experimentally measured surface deflection has been employed to solve the problem in this complex geometry.

The budget allotted for travel has been used for a one week visit of Dr. Dell'Aversana to Georgia Tech and to attend the American Physical Society conference in Kansas City, on March 20, 1997, in coincidence with a talk given by Prof. G. P. Neitzel to illustrate the first results of the Georgia Tech-Mars joint research (session O4: Surface-Tension-Driven Motion in Fluids and Solids). After his talk, Prof. Neitzel was invited by the Editor of the monthly scientific magazine "Physics Today" to contribute an article on this topic. During his stay at Georgia Tech, Dr. Dell'Aversana held a seminar on non-coalescence and non-wetting, helped a Prof. Neitzel's graduate student to set up the interferometer for a 2-D experiment on non-coalescence, collaborated with Prof. Neitzel to prepare his APS talk, and outlined, together with him, another presentation on the argument to be given at the Joint Xth European and VIth Russian Symposium on Physical Sciences in Microgravity, St. Petersburg, Russia, 15-21 June 1997. Such presentation is entitled "Features of non-wetting and non-coalescing drops" and is prepared by D. Castagnolo, P. Dell'Aversana, V. Tontodonato, and P. Neitzel.

References

[1]P. Dell'Aversana, J. R. Banavar and J. Koplik: "Suppression of coalescence by shear and temperature gradients," Phys. Fluids **8**, 15-28, (1996).

EXHIBIT A

Non-Coalescence Effects in Microgravity
(Subcontractor Part)

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Statement of Work and Budget Explanation
Year 1

According to the recommendations given in the final proposal review and in order to cope with the budget available, some changes have been operated with respect to the originally proposed study. The entire study will now concentrate on the sole coalescence topic, which is one of the two issues addressed in the original proposal. The MARS Center part of the work has been agreed with the Principal Investigator, Professor G. Paul Neitzel of the Georgia Institute of Technology in Atlanta. It will consist in an experimental activity, complementary to that to be carried out at Georgia Tech, and in a numerical simulation, to be conducted in collaboration with the group in America.

Experimental activity

The case where a non-coalescing system is surrounded by a gas will be investigated. The nature and the density of the gas are expected to affect the resistence of the interfaces between the non-coalescing liquids. In particular, we will see how the critical temperature difference between two liquids to hinder their coalescence depends on the outer gas pressure. An already existing cell will be adapted in order to change and monitor the gas pressure, to set the proper sample temperatures and to introduce the liquid and the gas indepedently from one another.

The interpretation of the stable non-coalesce effect in terms of elasto-hydrodynamic theory of lubrication can be experimentally backed by interferometric measurements which are able to reveal the presence, between the liquids, of a thin interstitial film of the external medium. Preliminary results in this sense have already been achieved at the MARS Center. In the course of this project, the experimental technique will be improved to determine the exact shape of the film in steady conditions. The results of these measurements will provide the surface contours to be used in the numerical simulations.

Numerical simulation

A numerical model for a non-coalescing system will be set out in collaboration with Georgia Tech using boundary conditions and parameter values which are consistent with the experiments. The disjoining pressure which we expect to find over the contact area will be used in cross checks with the experimental and theoretical results. The numerical results are also intended to help in determining the limits of validity of some assumptions and of the use of the Navier-Stokes equations to describe the flows within the thin film separating the non-coalescing liquids. This activity will be carried out in Italy: the original intent to have a researcher from Mars resident at Georgia Tech as a postdoctoral research associate, has had to be changed.

MARS Center Budget

Year 1

The budget share, also agreed with the Professor Neitzel, for the subcontract to the MARS Center for the first year of the project is \$ 35,083. In addition, one trip to Georgia Tech, of approximately 1 week duration is foreseen, whose cost has been estimated at \$ 2,500. About \$ 200 have been allotted for miscellaneous supplies, needed to support the experimental activity to be performed at MARS. The budget explanation is summarised below.

Subcontract to MARS	**\$ 35,083**
(\$ 88 / man-hour for year 1; 2.3 man-mo / yr)	
Travel	**\$ 2,500**
Miscellaneous Supplies	**\$ 200**
TOTAL FIRST-YEAR COST	**<u>\$ 37,783</u>**

EXHIBIT B

Suppression of Coalescence and of Wetting: the Shape of the Interstitial Film

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ABSTRACT

The shape of the interstitial air film between non-coalescing liquids and between a drop and a solid surface is detected by means of laser interferometry. Alternative optical techniques are exploited to reveal the film profile and absolute thickness and its dynamical behavior. In particular, the thickness of the film is measured, by means of an angle shift method, in the case of a drop resting on a solid surface; further experiments, based upon reflectometry, show that stable non-coalescence may be realised both under steady and time-dependent conditions of the interfaces. A light deflection technique, coupled with image analysis, is exploited to detect the features of the film unsteadiness. The related Fourier spectra are explicitly calculated. The procedure for the determination of the film thickness is explained with the aid of a specific numerical code which is able to simulate the interference fringe patterns generated by thin films. The results presented herewith are explained in terms of theory of lubrication, exploiting the experimental data as boundary conditions which allow one to uncouple the stress-balance equations at the interfaces and the lubrication equations and solve only the latter ones.

I. INTRODUCTION

In a paper that was previously published in this Journal[1] some cases were described where two liquids, separated by an air film, can be brought in close proximity of each other achieving a stable non-coalescence configuration. In that paper it was discussed how this phenomenon occurs in the presence of liquid surface motions, whereas coalescence would readily occur in the absence of such motions. It was also outlined that, no matter how the liquid surface motions are generated, it is possible to prevent the coalescence between two liquids provided that a threshold surface velocity (depending on the specific configuration) is overcome. The main hypothesis to explain this behavior was that the running liquid surfaces are able to drag air between the liquids so generating a constant lubrication contribution to the disjoining pressure. Thus, a permanent lubrication force would keep the two liquids at a distance where the van der Waals interaction may not be sufficient to drive the liquids in molecular contact and cause the primer of coalescence. In the present paper new experimental results, yielded by interferometric measurements, are presented that strongly support the hypothesis above. The present results definitively confirm the presence of the interstitial air film between the non-coalescing liquids. This film was already observed for the cases of temporary non-coalescence and its presence can now be generalised for the new liquid/liquid and liquid/solid systems here described.

As air is able to create a disjoining pressure between solids (as in common gas lubricated bearings) and between liquids (stable liquid/liquid non-coalescence) nothing prevents, in principle, the hindering of solid surface wetting by a liquid, if proper dynamical conditions are set. In fact, a qualitative result in this sense has been presented recently[2]. Generalising the term "self-acting bearing" used by Gross[3] in the classification of gas bearings, here we use the term 'self-lubricated systems' to indicate all the particular cases, including liquid/liquid and liquid/solid systems, where the gas lubricating film is supplied by the relative tangential motion rather than being supplied by an external source.

The study of self-lubricated systems can be relevant in a series of fields and in technological applications such as development of bearings and dampers, measurement of surface forces, rain formation, combustion efficiency of aerosols, composite materials, sticking of spray paints on smooth and clean surfaces, etc. Also, studies in rheology will benefit from the progresses in the understanding of the coalescence process and its inhibition[4].

Basic studies in gas lubrication may get ahead thanks to the powerful tool offered by laser interferometry gaining new insights on the features of lubricated systems in general and about their response to parameter variations in the lubricant.

In order to assess the relative importance of the lubrication contribution to the disjoining pressure, it is essential[5] to gain information about the gap shape and thickness. The relevance of these parameters is well known and has been confirmed by a number of experiments and theoretical calculations, many of which performed early in the Sixties[6, 7], with different configurations involving common, solid bearings. These studies have shown that small differences in the lubrication film shape and thickness imply dramatic differences in the ability of a lubricated system to support a load[8]. Laser interferometry offers a well suited means to gain deep insights on the gap shape and thickness.

Interferometry has already been used for the observation of the film separating two temporary non-coalescing liquids. It was exploited, for example, by Allan, Charles and Mason[9, 10] to reveal the film thinning that leads to coalescence of a bubble rising toward a liquid suface and the film thinning in the case of two immiscible layered liquids where a drop of the heavier liquid falls through the lighter liquid toward the liquid-liquid interface. Here, the novelty is represented by the fact that a refined interferometric technique has been applied to self-lubricated systems formed by a liquid drop against a solid surface. This configuration yields more accurate determinations of the film thickness and shape. In addition, the dynamical behavior of the interstitial film, which was still unknown, has been investigated for a liquid/liquid configuration. It is shown, in particular, how the air flows in the film may cease to be steady and give rise to periodic or aperiodic oscillations. This is reflected by the related unsteadiness in the film shape which starts changing in time even though the film does not undergo the rupture that would lead to coalescence. The unsteadiness in the film shape is observed here with special regard for the case of a silicone oil drop placed upon a bath of the same liquid.

In parallel with the experimental work presented herein, a specific numerical code has been developed to simulate the interference fringes produced by a thin film. It has been used here to support the interpretation of the interference patterns obtained in the experiments with air films.

II. EXPERIMENT DESCRIPTION

The experiments here described exploit thermocapillary convection (also referred to as thermal Marangoni convection)[11] to prevent coalescence. The thermal Marangoni effect is not the only way to achieve the non-coalescing and the non-wetting states[1, 12, 13]. The choice of exploiting thermocapillary convection is made because surface tension gradients can produce very regular surface flows which are relatively easy to set. Furthermore, thermal Marangoni convection is established without any external mechanical actuator, thus, the samples are free from vibrations that would compromise the interferometric observations.

Three different setups have been realised. The first one produces near-field fringes, which are focused on the thin film between a drop and a solid surface and are used to perform measurements of thickness; the second setup gives far-field fringes that are projected onto a screen and yield direct information on the main curvatures of the involved surfaces and on their behavior in unsteady situations; the third experimental configuration is used to detect the drop lateral surface oscillations exploiting a beam deflection technique coupled with image analysis, to yield indirect measurements of the air film oscillations.

A. Measurement of shape and thickness

The shape of the lubricating film between a drop and a reference flat surface of glass is revealed by means of near-field fringes. Fig. 1 depicts the interferometer used. One surface of the optical flat is machined with a flatness better than $\lambda/20$ (λ = 632.8 nm) and the opposite surface is treated with an antireflection coating. The drop is loaded upon the flat surface whilst the observation is made through the coated surface. The liquid/solid configuration is the best way to observe the shape of the lubrication channel because other configurations would complicate the interpretation of the fringe patterns. This would be, for example, the case where a drop is pressed against a curved solid surface, or the case of a drop pressed against the surface of another liquid. When the drop is pressed against a flat and rigid surface, the flat boundary of the film is used as a reference and the thickness gradients revealed by the interferometric patterns give complete information about the film profile, no other data being required except the absolute film thickness in one point.

In order to know the local film absolute thickness, the angle of incidence of the light was varied to observe the corresponding variation of the interference order at a given point. The thicker the film, the larger the interference order variation for a given angle shift. Fig. 2 resumes the situation. It sketches the Marangoni flows inside the drop and the velocity profile induced in the air film. Besides, the figure illustrates the variation of the light direction used to measure the film thickness.

Other techniques are available to measure the absolute thickness of a thin film: Newton's rings in white light give the approximate thickness from classical tables with the color successions; sophisticated heterodyne techniques[14] yield more precise measurements, but at the price of more complex experimental setups; ellipsometry is more appropriate for the nanometric range[15]. All in all, for our purposes, the angle shift method seemed to be the best compromise between precision and simplicity.

In the measurements discussed here, the angle of incidence was varied by linearly translating the laser head over the range l, as shown in Fig. 1.

Let λ be the wavelength of the incident light (a green HeNe laser was used at 543.5 nm), n_1 the refraction index of glass, n_0 the refraction index of air, d the unknown thickness, and Δm the interference order variation to be detected. The relation between Δm and d is thus given by:

$$\Delta m\lambda = 2dn_0\left[\sqrt{1-\left(\frac{n_1}{n_0}\right)^2\sin^2\alpha}-\sqrt{1-\left(\frac{n_1}{n_0}\right)^2\sin^2\beta}\right] \qquad (1)$$

where α and β are, respectively, the initial and the final angles of incidence of the light with respect to the normal of the film surface.

The angles α and β and the refraction indexes are known parameters of the experimental setup; thus, the quantity in square brackets in eq. (1) is a known constant, say A, resulting in the following:

$$d = \frac{\Delta m \lambda}{2 n_0 A} \qquad (2)$$

In order to avoid the risk of incorrectly estimating the thickness to be measured, the interferometer has been calibrated using a film of known thickness and of the same material (air) as that of the film to be measured.

The calibration consisted in measuring the interference order variation obtained in Newton's rings due to a spherical lens kept at a distance of 30.0 ±0.1 μm from the reference flat surface. So, equation (2) can be rewritten for the calibration distance d_0:

$$d_0 = \frac{\Delta m_0 \lambda}{2 n_0 A} \qquad (3)$$

where Δm_0 is the interference order variation related to d_0, obtained by translating the laser head over the range l. The value of Δm_0 measured for d_0 agreed with the expected one.

The angle shift range and all the remaining optical parameters were then left unchanged during the measurement of the unknown thickness. Thus, the thickness to be determined is readily obtained from equations (2) and (3) yielding:

$$d = d_0 \frac{\Delta m}{\Delta m_0} \qquad (4)$$

The technique illustrated above allows one to take advantage of the availability of steady conditions, being able to yield very precise measurements of thickness in the range 0.1 to 100 μm (for the wavelength used) with a spatial resolution approximately one order of magnitude better than that can be achievable with white light. Thanks to the use of monochromatic light, the contrast of the interference figures is enhanced and the analysis of the light intensity distribution is facilitated, without the need for any complex readout chain. Besides, precision can be further enhanced by means of image analysis techniques which are able to trace the light intensity in a well defined point of the interference figure.

The possibility to control the relevant parameters of the system under study - namely temperature gradients, volume of the liquids, pressure of the surrounding medium, and degree of liquid-to-liquid compression - without compromising their steadiness, allows one to monitor the evolution of the lubricating film features due to changes in the value of such parameters. The present paper reports the changes in the film shape produced by increasing the compression of a drop against a flat surface of glass.

Fig. 3 represents the result of the interferometric analysis as obtained for three different deformations of a drop of 5 cSt silicone oil on the flat reference surface mentioned above. In the three cases the volume of the liquid is kept unchanged while the position of the rod sustaining the drop is varied. The diameter of the rod is 5.0 mm; a constant temperature difference of 35 °C is imposed between the rod and the glass to establish a thermal Marangoni convection in the drop sufficient to ensure stable non-wetting (the rod is kept at a temperature of 54 °C while the temperature of the glass surface is 19 °C). The first thing to note is that the channel is not parallel and flat: rather, a dimple is present with an almost perfect axis symmetry, reflecting the axis symmetry both of the drop and the convective circulation which drives the lubricating air into the gap. This fact extends to the stable non-coalescence case the observations by Allan, Charles and Masons[9, 10] performed for the transient situations. One can observe that. as the drop is pressed against the glass and the contact area increases, the dimple deepens and the exit constriction reduces. Such deformations are necessary to balance the increased load. The drop deformation as a function of compression is shown in Fig. 4.

A numerical code has been developed to support the interpretation of interferometric patterns due to thin films of whatever refraction index and shape. Fig. 5 and Fig. 6 have been obtained with this code. using the film profile of Fig. 4, case b as input, to give a more immediate perception of a typical interference order variation found in the experiments. From the figures it is understood how the angle shift method described above can be used for the phase unwrapping of the interference patterns. Provided that the angle variation is performed quickly enough. the method can also be used in non-stationary situations, where changes in film shapes are slow with respect to the changes in the light direction.

In passing, it is important to note that the stable non-wetting state, in the presence of gravity, was observed only when the direction of the thermal gradient and the relative position of the drop and the solid surface was set in a well defined way: namely, it is necessary that the drop be warmer than the glass and stay above the glass. These observations constitute a further confirmation of the dragging action exerted by the moving liquid surfaces on the surrounding air. In fact, surface tension gradients have to drive the surface flows (and thus air flows) into the gap rather than outside. Moreover, since on ground, in silicone oil, buoyancy convection prevails over thermocapillary convection, the drop has to stay above the glass because otherwise, even with the drop warmer than the solid surface, one would have a flow rising along the drop axis and going down along the drop surface, thus dragging air out of the gap.

So far, the drop pressed against the flat glass surface has been referred to as 'non-wetting'. However, it is necessary to report that it is very difficult to achieve this state unless the drop is kept in close proximity of the glass surface for a while, before being pressed against it. In other words, if a clean and dry glass surface is used, then wetting almost always occurs quickly. On the other hand, if the hot drop is placed very near to the cold surface for a minute or so, a thin liquid layer is seen to slowly form on the surface itself, likely constituted by the liquid evaporated from the drop and subsequently condensed on the solid surface. Interferometry has shown that even cleaning the glass with a dry absorbing paper may not be sufficient to completely remove the thin oily layer which is clearly visible from the interference fringes it generates. If the drop is left in this position for enough time, then it is even possible to see droplets of oil migrating on the glass surface radially, moving away from the suspended drop vertex. Such migration is likely due to surface tension gradients and to the air flows which are present in the gap separating the drop from the glass. In some cases, one of these small droplets which condensed on the glass was also observed to remain trapped in the dimple formed after the suspended drop had been pressed against the glass. For these reasons one should be prudent when referring to 'non-wetting' droplets. However, we will continue to use this term because it is most expressive to refer to such particular self-lubricated systems.

B. Measurement of the air film oscillations

As shown below, stable non-coalescence can be achieved both as a steady and as an unsteady condition. In drop-bath systems, the thin lubricating film is strongly curved. Fig. 7 shows a situation of stable non-coalescence between a drop of silicone oil and a bath of the same liquid and offers a striking evidence of the possible interface deformations that can be achieved. In addition, the film shape may lose its axis symmetry and change in time.

When dealing with highly deformed interfaces, it is very difficult to determine the thickness gradient of the interstitial film for the whole extention of the film itself by means of near-field fringes. In fact, in such cases, only a small portion of the interference pattern is visible at a time, since the angle of incidence of light on the surface changes from point to point. In these situations far-field fringes in reflected light may help in giving other types of information. For example, from the deformation of the interference pattern one can get the curvature of the interfaces by means of triangulation. If the shape of the boundary interfaces of the thin film changes in time, far-field fringes are the only way to monitor the thickness variation over large portions of the film, even though the contrast of the figure may not be as good as for the near-field fringe patterns.

Fig. 8 represents a sketch of the setup used to obtain far-field fringes. The liquid of the bath is contained in a transparent cell to allow the laser light to be transmitted to the film. The beam splitter in the sketch deflects the laser beam toward the contact interfaces and transmits the interference figure toward a screen. The images of the fringes are recorded by a CCD camera as in the previous setup.

The bath container is fixed; the rod where the drop is suspended can be displaced along its axis, and the drop volume can be varied by means of an injection/suction pneumatic system. Thus the film shape can be monitored as a function of the drop position, volume, and temperature.

The disk sustaining the drop in Fig. 7 has a diameter of 5.0 mm and the drop is initially kept at a temperature of about 35 °C and at a constant volume while the bath is at ambient temperature. Subsequently, the temperature is raised to 70 °C and also its volume is increased. The unusual size of the drop shown in Fig. 7d can be achieved thanks to the hydrostatic push exerted by the bath which balances the weight of the drop. In the conditions of Fig. 7a the non-coalescing state is still steady. A view of the far-field interference pattern due to the lubricating film in steady conditions is represented in Fig. 9. Unsteadiness can be reached, for a given volume and shape of the drop, if the temperature difference with the bath overcomes a certain threshold. In Fig. 7d the unsteadiness threshold was largely overcome. This threshold depends on several parameters including the drop volume, the disk diameter, the viscosity of both the drop and the bath, the disk distance from the bath, the scale factor of the system, the absolute values of the involved temperatures (drop, bath, ambient). The conditions to reach the unsteadiness will not be analyzed in the present framework. Rather, it will be shown that the oscillations of the lubricating film can be periodic, provided that suitable conditions are set. Fig. 10 shows four moments in succession of a periodic oscillation in a lubricating film between a bath and a large drop whose diameter was 10.0 mm.

A dedicated experiment has been performed to measure the oscillations which affect both the lateral surface of the drop and the air film between the non coalescing bodies when the unsteadiness threshold is overcome. The question that one needs to address is whether the observed instability arises in the drop or in the film. In facts, while it seems reasonable that wide gas films may give rise to instabilities, another possibility is that the oscillations observed in non-coalescing drops arise in the drops themselves, as a result of capillary convection instabilities of the same nature as those encountered in liquid bridges[16, 17]. In this respect, such drops might be regarded as liquid bridges whose bottom lays on the colder bath surface.

The behavior of the drop oscillations has been observed as a function of the temperature difference between the disk sustaining the drop and the bath. In order to monitor such oscillations in various experimental conditions, again, a non-invasive optical technique has been used. The technique consists of exploiting a properly focused laser beam which, being transmitted through the drop, projects a light spot on a screen. The light spot position on the screen is very sensitive to any liquid surface deformation. The image of the light spot is acquired by a frame grabber and digitised. The position of the spot centroid is then automatically calculated and tracked for about 40 seconds. The experimental setup is sketched in Fig. 11; the data collected in this way for three different temperature conditions above the unsteadiness threshold (about 25 °C, in this event), are shown in Fig. 12 together with the related frequency spectra, obtained by means of a FFT algorithm. Here the disk sustaining the drop has a diameter of 5.0 mm.

It can be noted that the main frequency peak moves toward the high frequencies as the temperature difference is increased. Simultaneously, the oscillation amplitude also increases. Such a behavior with respect to the ΔT variations is similar to that found in liquid bridges[18] and, if one considers that also the frequency value of the main peak is of the same order of magnitude as that of liquid bridges, then it seems reasonable to suppose that the observed unsteadiness starts in the drop and is subsequentely transmitted to the air film through the oscillations of the liquid velocity at the surface.

The oscillation frequency of the air film has also been desumed directly, from an analysis of the interference pattern dynamics recorded with the setup of Fig.6. Due to the larger complexity of the optical image to be analysed, with respect to the light spot, broader frequency spectra are yielded by these measurements, but with identical main peak values indicating the coupling of the two oscillations (see Fig. 12c).

Even though these experiments seem to indicate that the unsteadiness observed is in reality nothing different from a floating zone instability, the present data cannot exclude that the instability arises in the film. This subject should be further investigated, e.g., by means of linear stability theory[19, 20].

The observations made indicate that the same type of instability occurs also in the drop-glass system, even if the amplitude of the oscillations was much less in this case.

III. DISCUSSION OF THE EXPERIMENTAL RESULTS

In the following some arguments are presented to discuss the dimpled film profiles found experimentally and to show how the lubrication action of the air film gives rise to non-coalescence and non-wetting. The discussion will be based on the non-wetting of a glass surface by a silicone oil drop, as the experimental data on the film profiles given herein refer to such configuration, but similar considerations apply also to non-coalescence of liquids, where an analogous dimpled film is formed as well.

In principle, the description of the phenomenon should be based on the energy-momentum balance equations, coupled with mass conservation and with boundary conditions at the interfaces, but in practice the lubrication equations to be used in this specific problem (where small loads and Reynolds numbers are involved and where friction is by far smaller than in common bearings), can be obtained with some simplifications, neglecting the energy relations[3] and starting from the well known Navier-Stokes equation of momentum conservation:

$$\rho\frac{\partial \mathbf{v}}{\partial t}+\rho(\mathbf{v}\cdot\nabla)\mathbf{v}=\nabla\left[(\mu+\mu^*)\nabla\cdot\mathbf{v}-p\right]+\mu\nabla^2\mathbf{v}+\mathbf{F} \tag{5}$$

where ρ is the density, $\mathbf{v}$ is the fluid velocity, μ and μ^* are the shear and the bulk viscosity respectively, p is the absolute pressure and $\mathbf{F}$ represents the body forces.

Let a case be considered where the air film is steady (like in Figs. 3 and 4, for example). This corresponds to a situation where also the air flows within the film are steady. The air film can be safely assumed to be adiabatic, because the thermal conductivities of the liquid and of the glass are much larger than that of air. As in many cases of gas lubricating films, the air film is considered incompressible, and the viscosity μ uniform throughout the channel. In the present situation these assumptions can be made, since the involved loads are quite small (some dynes) and practically no heat is produced inside the film; in addition, the body forces can be neglected (the hindrance of coalescence has been produced in microgravity as well[1]). Considering the problem in cylindrical coordinates and expressing the air velocity $\mathbf{v}$ as

$\mathbf{v} = u\mathbf{i}_r + v\mathbf{i}_\vartheta + w\mathbf{i}_z$, imposing the axis symmetry of the problem, one can write the two surviving components of eq. (5) as:

$$\rho\left(u\frac{\partial u}{\partial r} + w\frac{\partial u}{\partial z}\right) = -\frac{\partial p}{\partial r} + \mu\left(\frac{\partial^2 u}{\partial r^2} + \frac{1}{r}\frac{\partial u}{\partial r} - \frac{u}{r^2} + \frac{\partial^2 u}{\partial z^2}\right) \qquad \textit{radial component}$$
$$\rho\left(u\frac{\partial w}{\partial r} + w\frac{\partial w}{\partial z}\right) = -\frac{\partial p}{\partial z} + \mu\left(\frac{\partial^2 w}{\partial r^2} + \frac{1}{r}\frac{\partial w}{\partial r} + \frac{\partial^2 w}{\partial z^2}\right) \qquad \textit{axial component} \qquad (6)$$

A further simplification can be made observing that the axial component of the air velocity is, in average, small, compared with the radial one (to complete its path *across* the film, a certain mass of air needs to go all the way forth and back *along* the film radius and the film radius is two orders of magnitude larger than the film thickness).

Therefore eqs. (6) reduce to:

$$\rho u\frac{\partial u}{\partial r} = -\frac{\partial p}{\partial r} + \mu\left(\frac{\partial^2 u}{\partial r^2} + \frac{1}{r}\frac{\partial u}{\partial r} - \frac{u}{r^2} + \frac{\partial^2 u}{\partial z^2}\right)$$
$$\frac{\partial p}{\partial z} = 0 \qquad (7)$$

Thus, as a consequence of the approximation just made, one gets that the pressure within the film does not depend on z.

At this stage, further order of magnitude considerations can be made operating the following normalizations:

$$\bar{p} = \frac{pH}{\mu V_M}; \qquad \bar{u} = \frac{u}{V_M}; \qquad \bar{r} = \frac{r}{R}; \qquad \bar{z} = \frac{z}{H}$$

and the aspect ratio $A = H/R$ and the Reynolds number $\mathrm{Re} = \rho V_M H/\mu$ are introduced, V_M being the reference liquid surface velocity, R the channel radius, and H the reference channel height.

Now, the first of eqs. (7) can be rewritten in a non-dimensional form:

$$A\bar{u}\frac{\partial \bar{u}}{\partial \bar{r}} = -\frac{A}{\mathrm{Re}}\frac{\partial \bar{p}}{\partial \bar{r}} + \frac{A^2}{\mathrm{Re}}\left(\frac{\partial^2 \bar{u}}{\partial \bar{r}^2} + \frac{1}{\bar{r}}\frac{\partial \bar{u}}{\partial \bar{r}} - \frac{\bar{u}}{\bar{r}^2} + \frac{1}{A^2}\frac{\partial^2 \bar{u}}{\partial z^2}\right) \tag{8}$$

In order to assess the relative importance of the various terms in the equation above, the typical values of the experiments described in the text are assigned, namely: $\rho = 1.2\cdot 10^{-3}$ g/cm^3; $\mu = 1.81\cdot 10^{-3}$ poise; $V_M = 0.5$ cm/sec; $H = 10^{-3}$ cm; $R = 5\cdot 10^{-2}$ cm. With these values $A = 2\cdot 10^{-2}$ and $\mathrm{Re} \approx 3.3\cdot 10^{-4} \approx A^2$. So, neglecting all the terms of order $\geq A^0$, and transposing equation (8) to its original dimensional form one can write:

$$\frac{\partial p}{\partial r} = \mu\frac{\partial^2 u}{\partial z^2} \tag{9}$$

Integrating eq. (9) twice across the film profile yields a parabolic Poiseuille profile for the velocity u, as anticipated in Fig. 2:

$$u = az^2 + bz + c \tag{10}$$

The three coefficients in the expression of the velocity are determined imposing the constraints of the problem. They are: $u(z = 0) = 0$ and $u(z = h(r)) = U_b(r)$, assuming no-slip at the film interfaces, $U_b(r)$ being the liquid surface velocity (which is not constant in r); $\int u(r, z)dz = 0$, which states that the net flux along the lubrication channel vanishes, contrary to most situations, where the lubricant does not undergo a velocity inversion across the channel. As far as the no-slip assumption is concerned, it can be observed that it is verified in the present circumstances, as the Knudsen number results to be $< 10^{-2}$, with the typical values which characterize the problem ($Kn = \lambda/H$, λ being the mean free molecular path of air molecules which is equal to $\approx 6.3\cdot 10^{-2}$ μm at normal ambient conditions).

With the constraints introduced above, eq. (10) assumes its explicit form:

$$u = u(r,z) = \frac{3U_b(r)}{h^2(r)} z^2 - \frac{2U_b(r)}{h(r)} z \tag{11}$$

and, together with eq. (9), makes it possible to derive the following expression:

$$\frac{\partial p}{\partial r} = 6\mu \frac{U_b(r)}{h^2(r)} \tag{12}$$

which needs to be integrated to obtain the pressure profile within the lubricating air film.

h(r) has been measured in some cases and, as reported in the previous pages, it can be described with good approximation by a fourth order, polynomial function; for $U_b(r)$ precise data are not available yet (its maximum value has been measured to be around 0.5 cm/s), nevertheless, it can be noted that it must be a monotonic funtion of r, which vanishes at the origin.

As a result of eq. (12), after integration, one finds out that, should h be constant in r, the overpressure, generated inside the channel by the incoming flow, would be maximum at the film center. This overpressure, being the liquid readily deformable, produces a local surface displacement. This, in simple terms, is how the dimple forms. Also the counterintuitive deepening of the dimple revealed by the measurements, occurring when the drop is squeezed against the glass, has a qualitative explanation. As the drop is pressed against the rigid, flat surface, in the absence of wetting, it undergoes an elastic flattening with consequent widening of the film area. Therefore the pressure, even if one assumes that its value does not increase, acting over a larger surface, produces a deeper deformation.

It is understood that, in order to calculate the pressure curve accounting for the surface deformations, one should work with the stress-balance equations at the air/liquid interface and with fluid-dynamic equations simultaneously. That would be a rather difficult task, considering that the deformation amplitude depends on the local dynamical situation at the liquid surface, which is undergoing convection due to temperature - and, thus, surface tension

- gradients. Fortunately, the film shape is an experimental data, and this allows one to just deal with the equation of momentum inside the lubrication channel.

The steady conditions of the present example are conditions of dynamical equilibrium where the Laplace's force generated by the liquid surface deformation opposes the dynamical pressure exerted by the air, which tends to further deform the channel.

One can thus conclude that the lubrication force supplies a disjoining contribution to the total force acting on the involved interfaces. In the absence of such disjoining contribution due to lubrication, the surface attractive forces, not contrasted, would lead the system to wetting (or coalescence, according to the relevant case).

IV. CONCLUSIONS

The present work shows that the phenomena of the hindrance of coalescence and of wetting can be explained in terms of lubrication and that some results found by other authors[9, 10] in the past for temporary non-coalescing bodies can be generalised to stable non-coalescing and non-wetting systems. Namely, as in temporary non-coalescence, the systems investigated here are actually separated by a thin film of the surrounding medium and this film is not flat but it is thicker at its center. The need to measure with precision the characteristic shape of this film is understood in the light of elasto-hydrodynamic theory of lubrication, from which it is known that even small variations in the film profile may yield important differences in the pressure curves and thus in the ability of the system to sustain a load. An attempt to calculate such pressure curves between non-coalescing liquids has been recently made[2] by means of numerical calculations which assume a parallel and flat interstitial film. Now, more precise results can be obtained, accounting for the actual shape of the channel, which has been detected by means of laser interferometry under different experimental conditions.

Near-field fringes have been used to measure the absolute thickness and the shape of the interstitial film of a liquid/solid configuration. The technique is based upon an angle shift method and has been illustrated with the aid of numerical simulations of the interferometric patterns. The experimental data, yielded by such measurements, supply the boundary conditions to solve the lubrication equations, uncoupling them from the interface stress-balance equations, provided that the velocity distribution $U_b(r)$ at the liquid surface is known.

Measurements made both with far-field fringes and by a beam deflection technique on liquid/liquid configurations have yielded quantitative data evidencing that such self-lubricated systems may undergo unsteady behavior when the values of parameters such as, e. g., temperature or volume, overcome a certain threshold. The unsteadiness can be periodic and does not necessarily lead to the interface rupture. The oscillatory behavior of the systems in question has been suspected to be of the same nature as that of the thermocapillary convection instability in floating zones. Nevertheless, the present data cannot exclude that the observed unsteadiness arises in the lubricating film itself and is subsequently transmitted to the drop bulk. A possibility to clarify this point is to use, for example, the tools of the linear theory of instability.

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FOOTNOTES AND REFERENCES

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FIGURE CAPTIONS

FIG. 1. Sketch of the experimental setup used to reveal the shape and measure the thickness of the interstitial film between a silicone oil drop and a reference surface of glass.

FIG. 2. When the drop is hotter than the solid surface a Marangoni convection develops in the drop. The surface flow is directed toward the colder part and it drags an air film (not to scale in the drawing).

In order to measure the air film thickness, the incident light direction is changed from α to β to observe the corresponding interference order variation, which is proportional to the local film thickness.

FIG. 3. Drop of 5 cSt silicone oil pressed against a flat glass surface. In row 1 the drop has been pressed by 100 μm with respect to the position where the first interference fringes appear; in row 2 the displacement is 200 μm; and in row 3 it is 300 μm. The three columns represent respectively:

a) the drop on the reference surface;

b) the near-field fringes due to the thin air film between the glass and the drop;

c) a 3-D view of the lubricating film shapes as reconstructed from the interference patterns.

FIG. 4. Comparison of the film profiles obtained for the three different compressions of the drop against the glass surface as shown in Fig 4.

a) 100 μm displacement

b) 200 μm displacement

c) 300 μm displacement

FIG. 5. The near-field interference fringes obtained from a numerical simulation using the interstitial film profile of Fig. 4b for different incidence angles of illumination. From the frame sequence it is possible to discriminate minima and maxima since the interference order variation, for a given angle shift, is larger where the film is thicker.

FIG. 6. Plot of the light intensity versus the illumination direction in correspondence of the maximum (central point) and minimum (external point) thickness of the interstitial film as obtained from the simulation. Similar plots can be obtained from the experiments and used to determine the interference order variation.

FIG. 7. Inhibition of coalescence between silicone oil drops of increasing volumes and a bath of the same liquid. Here, the disk sustaining the drop has a diameter of 5.0 mm, but larger sizes are possible, also in the presence of gravity. In frame d the disk has a temperature of 70 °C while the bath is at ambient temperature.

FIG. 8. Sketch of the experimental setup used to monitor the time-dependence of the interstitial film shape in unsteady conditions for a drop/bath self-lubricated system.

FIG. 9. The far-field fringes produced by the interstitial film of a drop/bath system as they appear in steady conditions. The drop diameter in this case is 10.0 mm.

FIG. 10. Sequence of far-field fringes produced by the same system as that of the previous figure, driven to unsteadiness. Note the apparent saddle like shape of the lubricating film. The numbers represent the time in hh:mm:ss.

FIG. 11. Setup used to track the drop lateral surface displacements. The data collected with this technique constitute an indirect measurement of the air film oscillations.

FIG. 12. Oscillation amplitudes (windowed) and related frequency spectra for three different temperatures of a 5 mm diameter drop: a) 47.3 °C; b) 51.8 °C; c) 56.0 °C. The bath temperature in all cases is fixed at about 20 °C. The broader spectrum shown in c) has been obtained by a direct analysis of the interference pattern rotation.

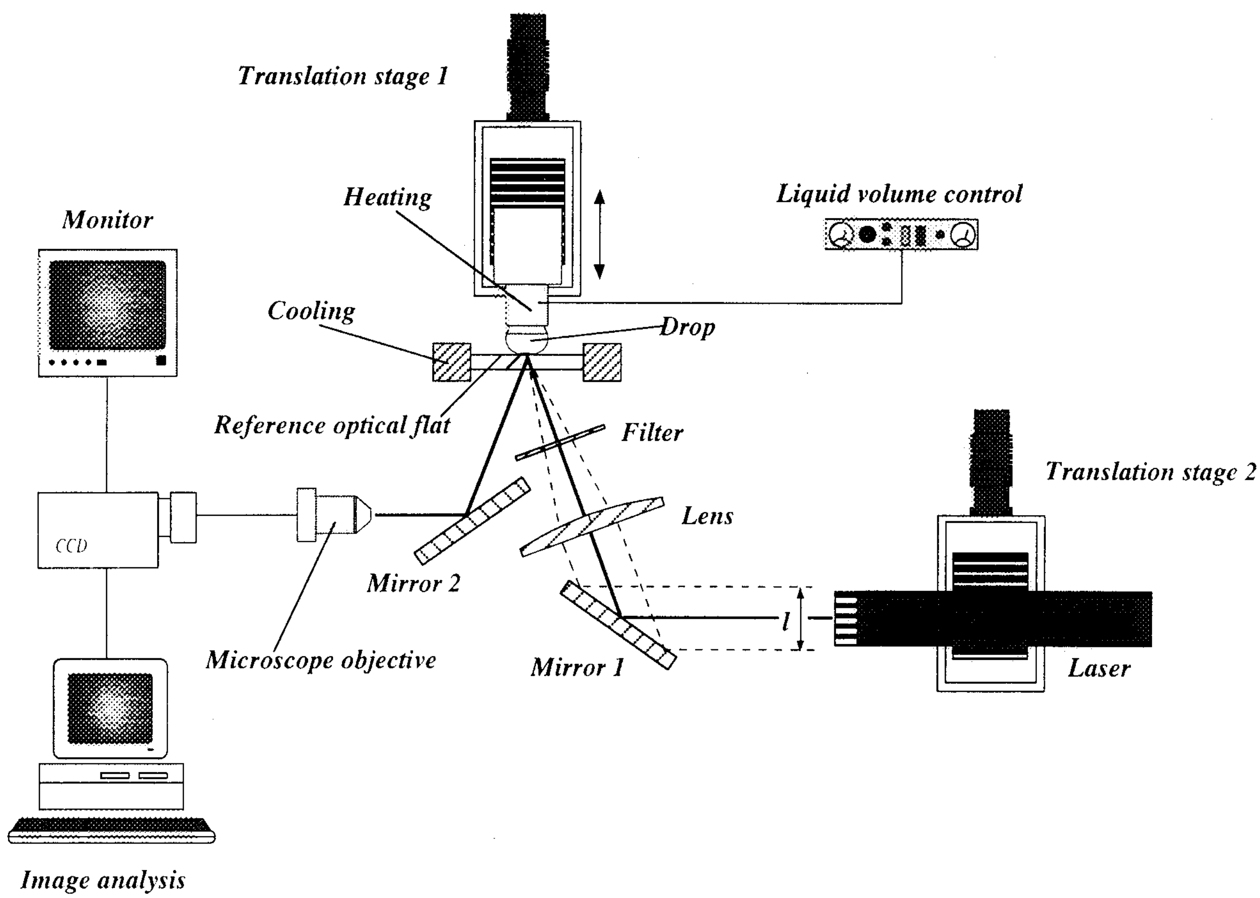

Translation stage 1
Heating
Liquid volume control
Monitor
Cooling
Drop
Reference optical flat
Filter
Translation stage 2
Lens
CCD
Mirror 2
l
Microscope objective
Mirror 1
Laser
Image analysis

Fig. 1

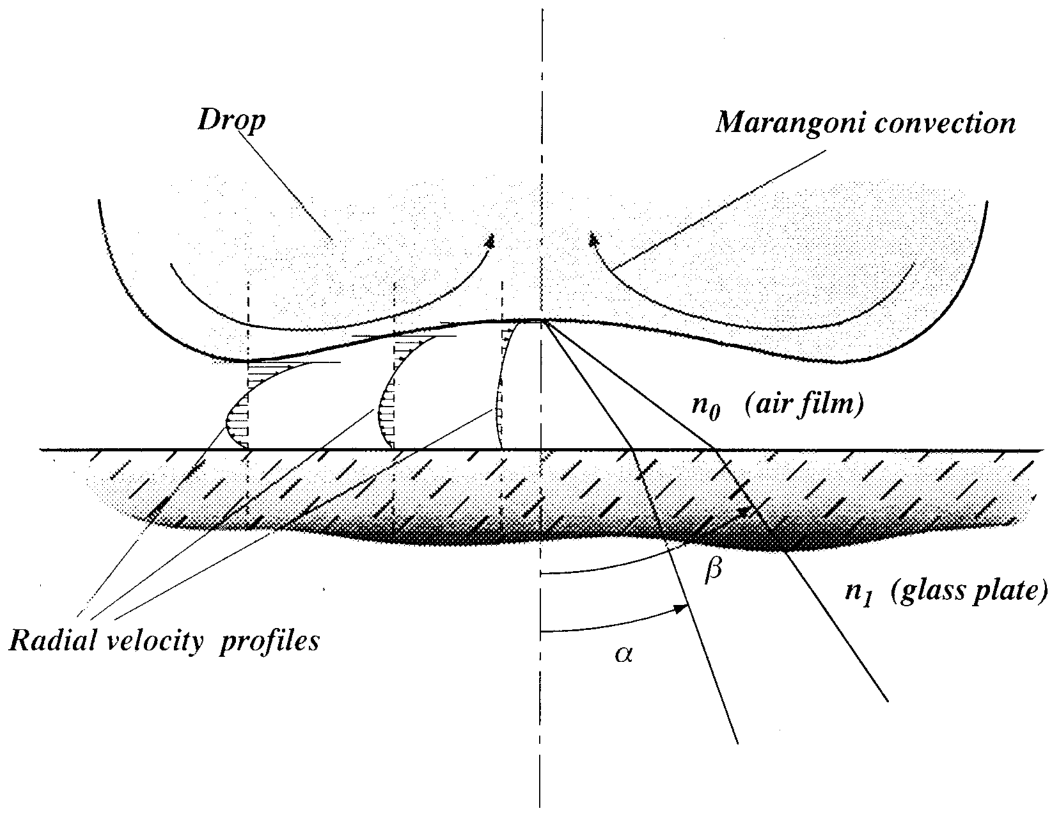
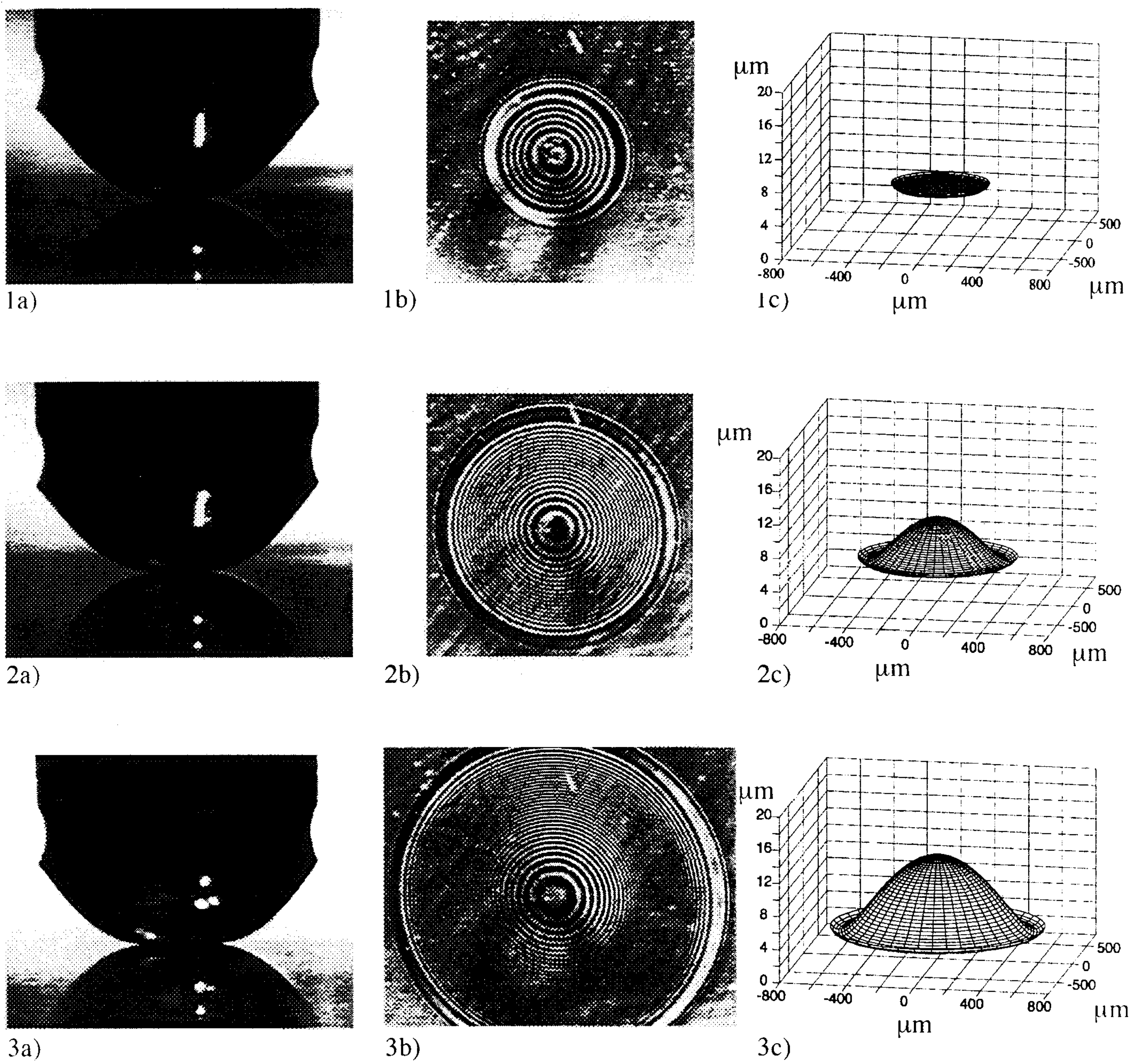

Drop
Marangoni convection
n_0 (air film)
β
n_1 (glass plate)
Radial velocity profiles
α

Fig. 2



1a)
1b)
μm
20
16
12
8
4
0
-800
-400
0
400
800
500
0
-500
μm
μm
1c)
2a)
2b)
2c)
3a)
3b)
3c)

Fig. 3

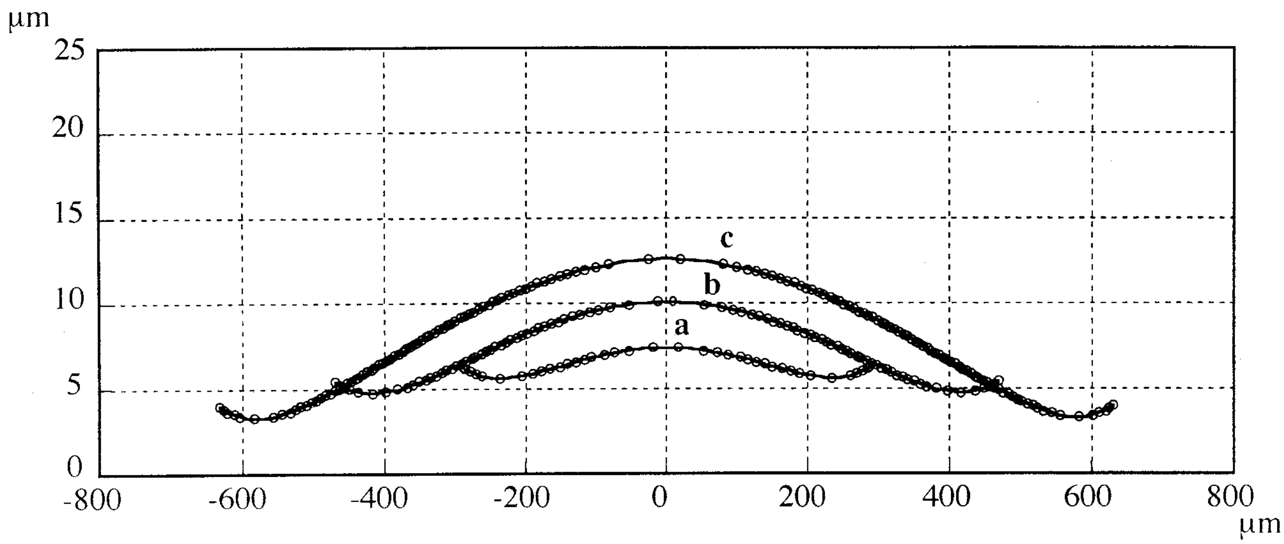


Fig. 4

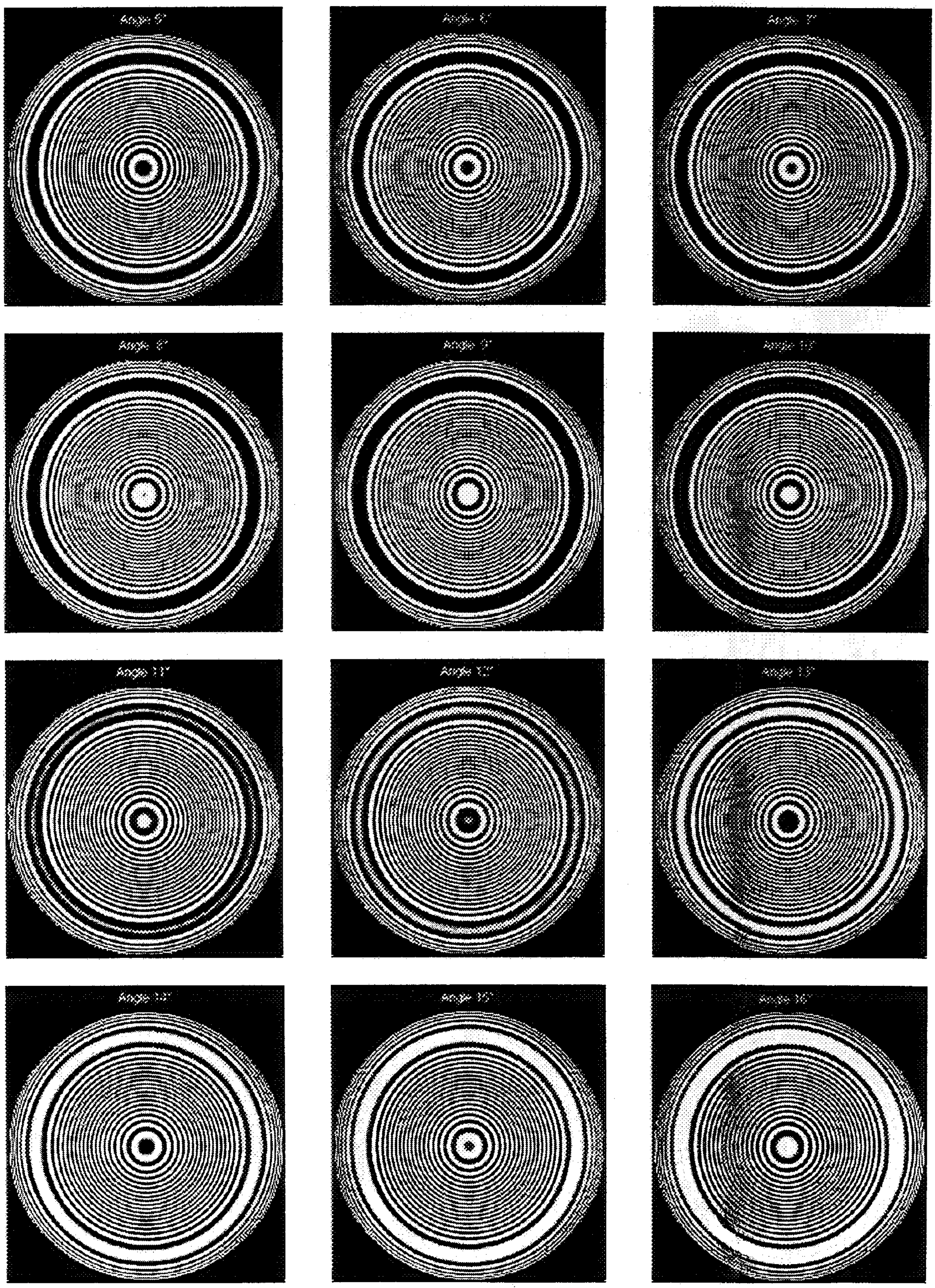
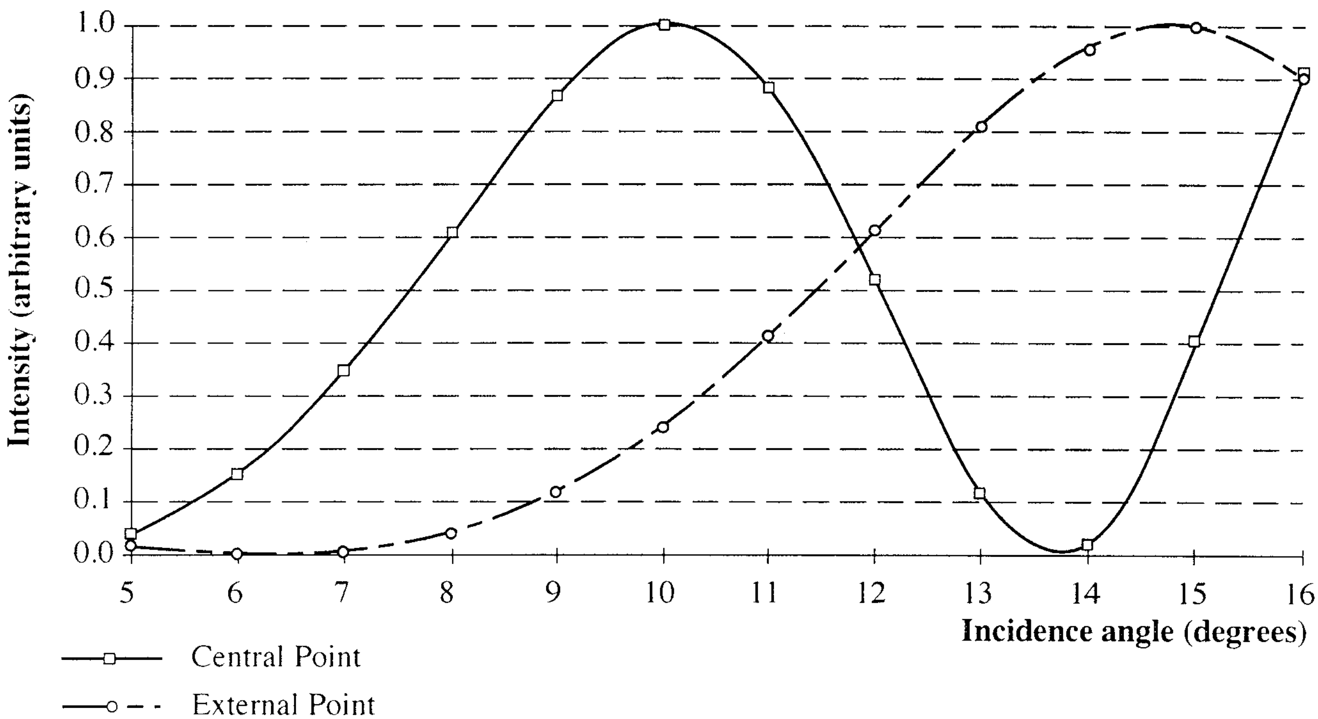


Fig. 5



1.0
0.9
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
0.0
Intensity (arbitrary units)
5
6
7
8
9
10
11
12
13
14
15
16
Incidence angle (degrees)
Central Point
External Point

Fig. 6

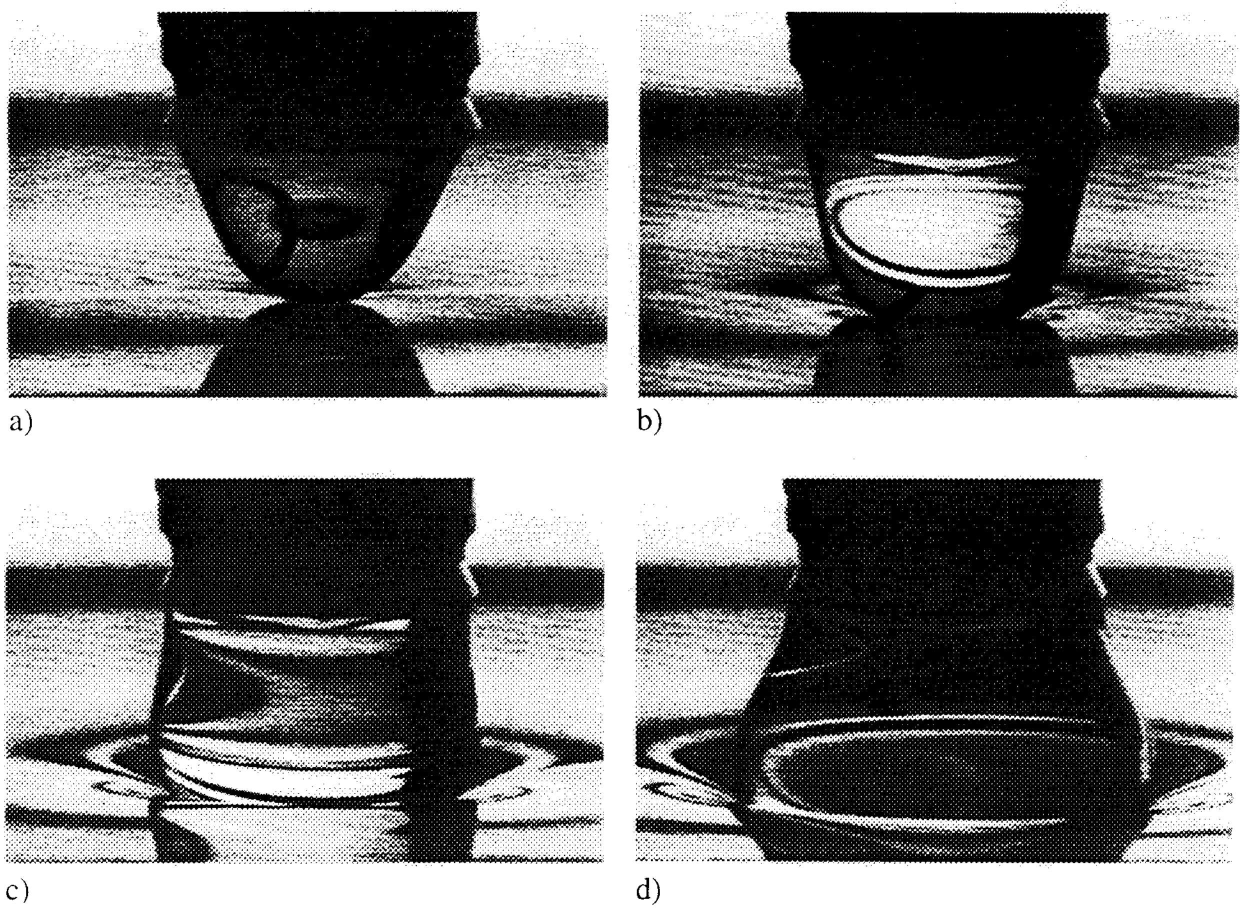


Fig. 7

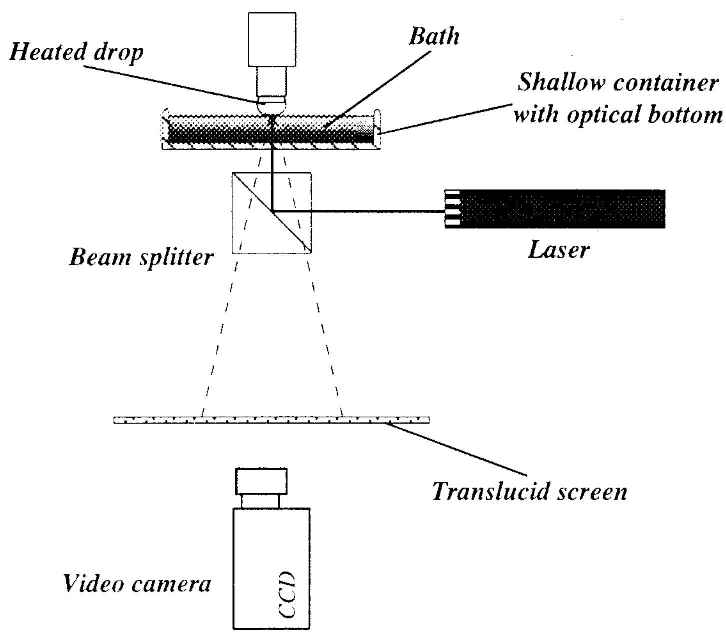

Heated drop
Bath
Shallow container
with optical bottom
Beam splitter
Laser
Translucid screen
Video camera
CCD

Fig. 8

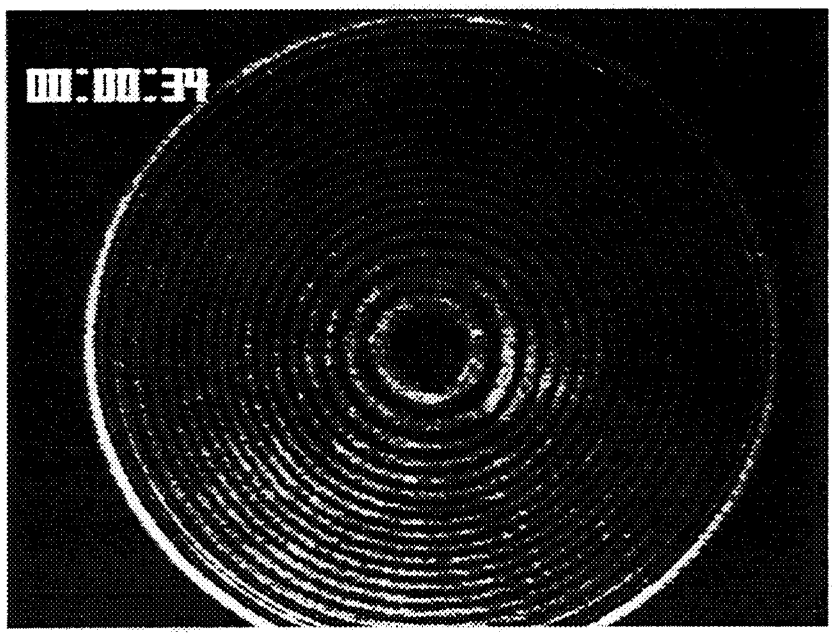
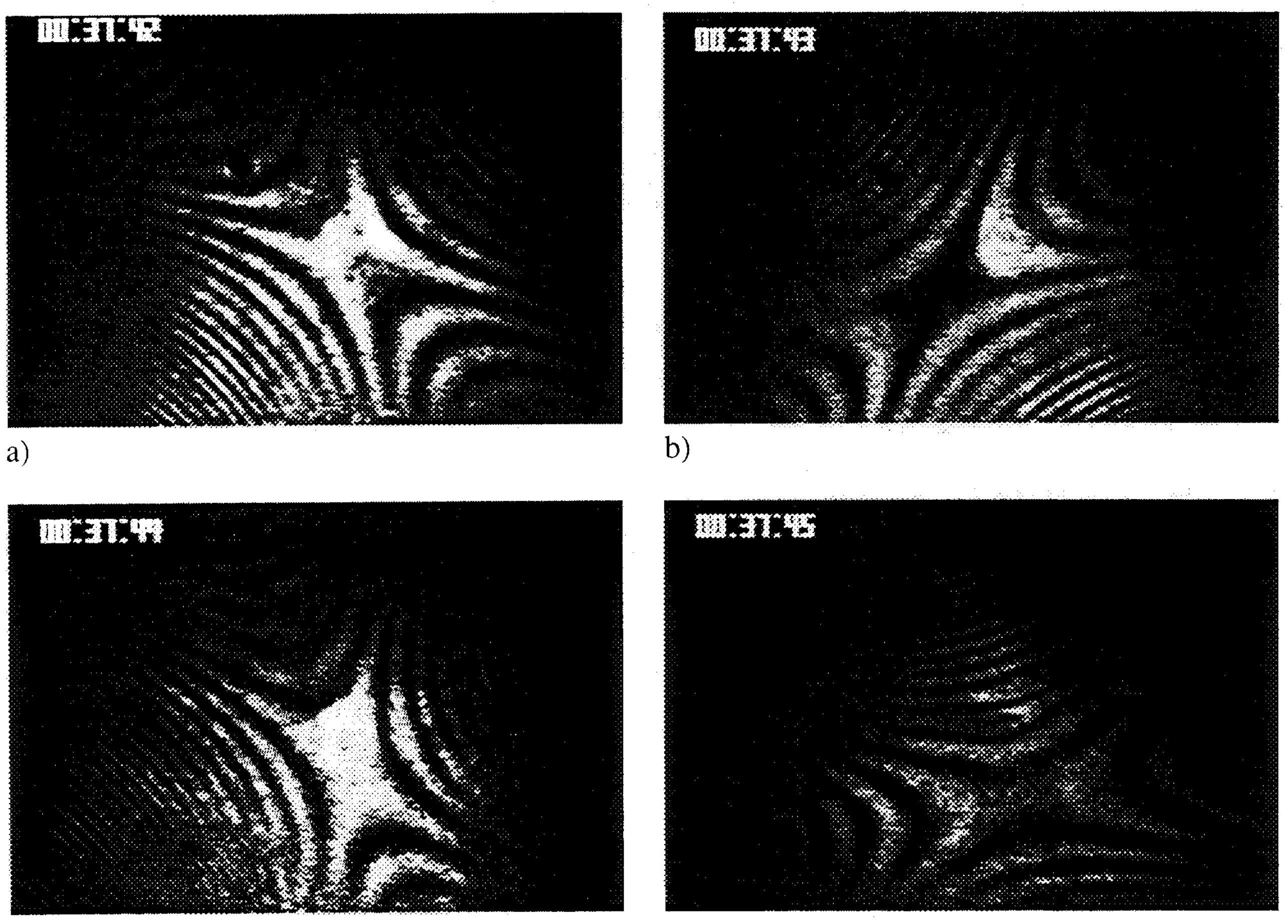


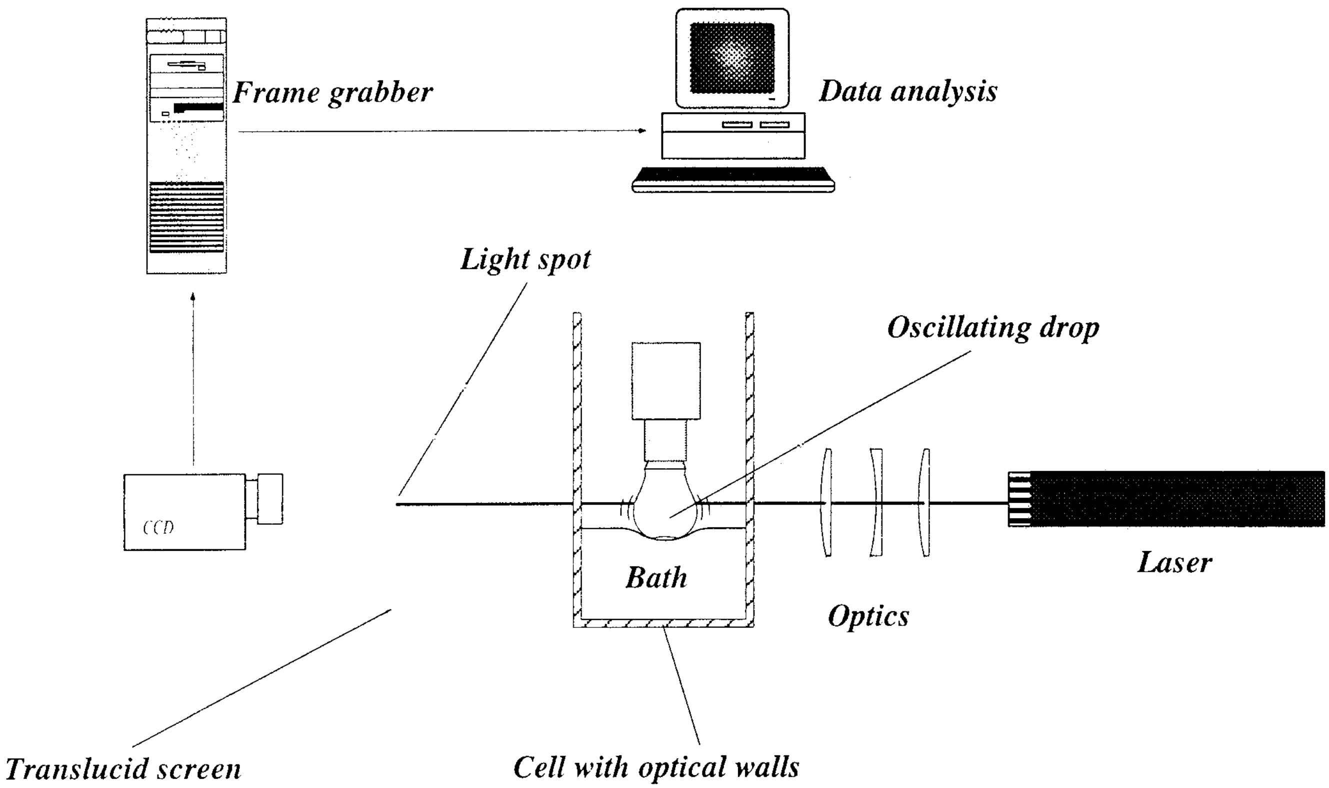
Fig. 9



a) b)

c) d)

Fig. 10



Frame grabber
Data analysis
Light spot
Oscillating drop
CCD
Bath
Laser
Optics
Translucid screen
Cell with optical walls

Fig. 11

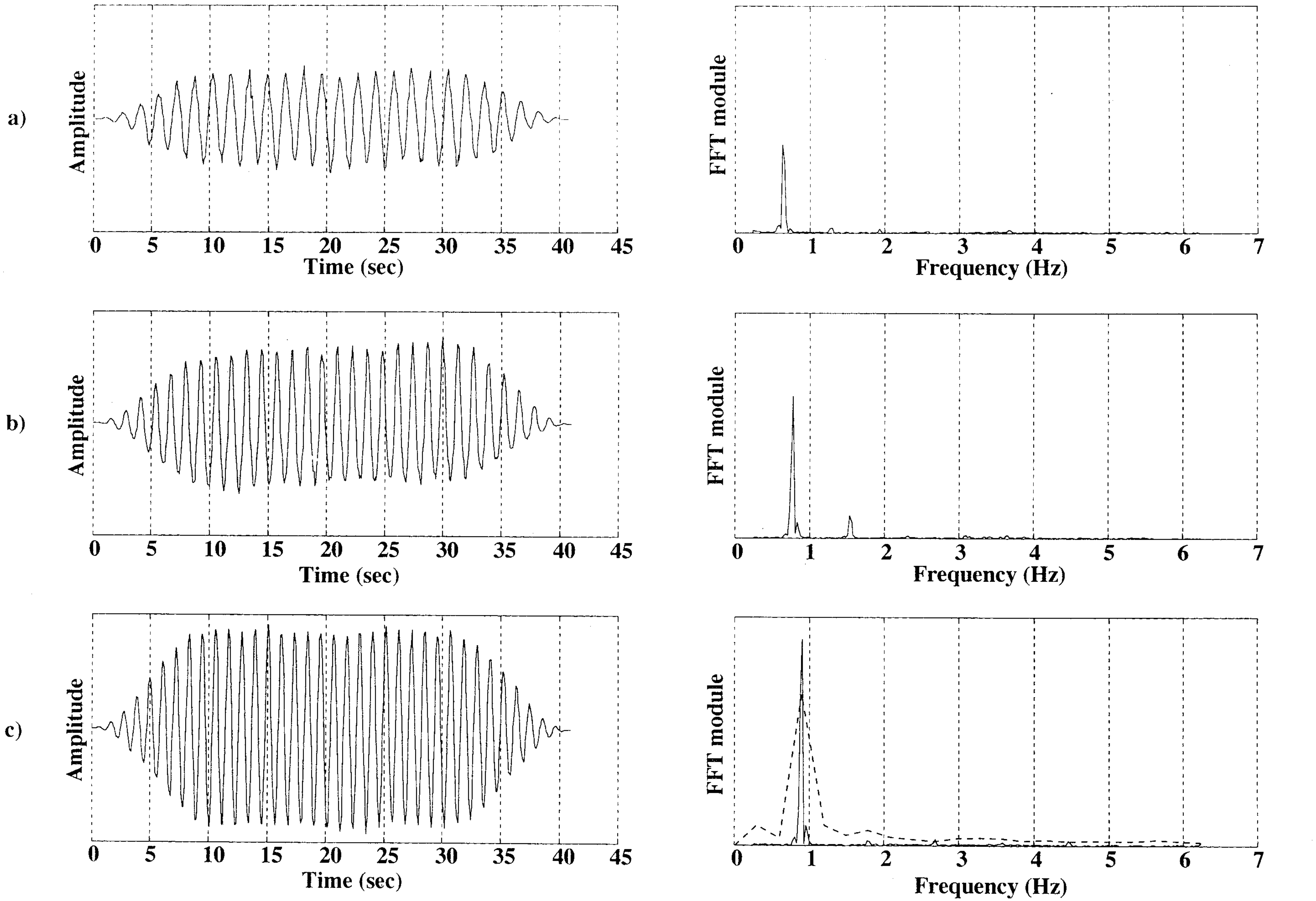

a)
b)
c)
Amplitude
Time (sec)
0 5 10 15 20 25 30 35 40 45
FFT module
Frequency (Hz)
0 1 2 3 4 5 6 7

Fig. 12